



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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First Named Inventor : Jobst LA DOUS
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Title : Photochromic Plastic Object

DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Jobst La Dous, hereby declare as follows:

1. I am a citizen of Germany, residing at Johansenaue 146, Krefeld, Germany 47809.

2. I studied ECONOMICS from 1970 to 1973 at FG HÖNCHENGRASSE and received the degree of DIPLOM in

3. Since 1979 I have been employed by KOVARJO as GENERAL MANAGER in the . My responsibilities include

4. I am an inventor of the present invention which is disclosed and claimed in the above-identified patent application, which relates to a photochromic synthetic resin body and a method of producing such a photochromic synthetic resin body, and I have read and understood the patent application. I make this Declaration in support of the patent application.

5. I have also read and understand Sommerfeld et al., U.S. Patent No. 5,886,101, which has been cited as evidence of the state of the art during the examination of the above-identified patent application.

6. The following experiments were carried out by me or under my direction and demonstrate that a resin body comprising at least two interpenetrating polymer networks and having a photochromic dye homogeneously distributed therein as claimed in the above-identified patent application displays enhanced photochromic response kinetics.

7. A first polymer network can be made of polyurea or polyurethane, and a second polymer network can be made of polyacrylate, polymethacrylate or a mixture thereof.

8. A photochromic dye can be homogeneously distributed within the synthetic resin body by introducing the dye to the interpenetrating polymer networks before or during polymerization.

9. A mass dyeing process can be used to distribute a photochromic dye throughout the interpenetrating polymer networks. In a mass dyeing process, a photochromic dye is added to the casting resin mixture prior to polymerization in the absence of a solvent. Solvents must be excluded from the mass dyeing process in order for the dye to be incorporated in the polymer network. In the presence of a solvent, photochromic dyes would remain in solution.

10. The following experiments demonstrate that the photochromic response (i.e., darkening and lightening behavior) of the claimed interpenetrating polymer network-based transparent synthetic resins is substantially enhanced with respect to conventional (comparative) synthetic resin materials.

11. Examples 1 and 2 describe the synthesis procedures for comparative photochromic synthetic resins.

12. Comparative Example 1 - pure polyacrylate:

Ten grams of monomer TS-150 (Tokuyama Company, 20-30% methacrylic acid ester, 70-80% polyethylene glycol dimethacrylate) were stirred with 2 milligrams of photochromic dye (spiro-9-fluoreno-13'-[3-[4-(N-piperidinyl)phenyl]-6-methoxy-3-phenyl-indeno[1,2-b]pyrane, Rodenstock GmbH) and 100 milligrams of tert-butylperoxyneodecanoate (TBPND, Peroxid Chemie). Polymerization was carried out at 30-85°C according to the polymerization protocol provided by Tokuyama Co.

13. Comparative Example 2 – pure polyurethane:

6.45 grams of polyetherpolyol (Desmophen®1262 BD, Bayer AG) was stirred with 2 milligrams of the photochromic dye used in Comparative Example 1 and 386 grams of isocyanate (Desmodur® 3400, Bayer). Polymerization was carried out at room temperature via the addition of 42 milligrams of bismuth catalyst (Octa-Soligen Bismuth 24, Borchers, GmbH). Final curing was carried out in an oven at 50°C for 30 minutes.

14. Examples 3 and 4 describe the synthesis procedures for photochromic synthetic resins according to the invention comprising at least two interpenetrating polymer networks.

15. Example 3 – Polyacrylate/polyurethane network:

By stirring together 50% each of the polyacrylate and polyurethane materials from comparative Examples 1 and 2, a predominately polyurethane network was formed over about one hour at room temperature. Subsequent polymerization of the acrylate was carried out in an oven at 30-85°C.

Note: by replacing the TS-150 with a more reactive acrylate monomer such as trimethylpropyltrimethacrylate (TMPTMA), interpenetrating networks can be formed concurrently without the addition of peroxide.

16. Example 4 – Polyacrylate/polyurea network:

To 4.2 grams of asparaginic acid ester (Desmophen®NH 1420 – difunctional, Bayer) were added:

2 milligrams of the photochromic dye used in Comparative Example 1;

2.7 grams of trifunctional isocyanate (LV 2, Rhodia Chemie);

3.0 grams of tetraethylene glycol diacrylate, and
1 drop of TBPND.

The cross-linking of both networks was carried out at room temperature as in Comparative Example 2, and then in an oven at 80-85°C for one hour.

17. The syntheses described in Examples 1-4 were carried out without the use of solvents.

18. The photochromic properties of test samples made from Examples 1-4 were evaluated in the laboratory. The test samples were prepared by pouring the photochromic mixtures from Examples 1-4 into a plastic ring mold (1.2 mm x 7 cm diameter) that is positioned on a glass sheet measuring 10 cm x 10 cm. The assembly was covered with a second glass sheet, and the sample was polymerized. The mold was dismantled and each test sample was measured using a kinetic bench according to DIN EN ISO 8980-3 at 23°C. Samples were irradiated with 50 klux (50,000 lux) for 15 minutes, and the lightening (inverse darkening) was monitored for an additional 15 minutes.

19. Darkening and lightening data of the comparative test samples (1 and 2) and samples according to the invention (3 and 4) are shown in Table 1.

Table 1. Darkening and Lightening Results

Property (%)		Comparative Examples		Inventive Examples	
		1	2	3	4
i.	Transmission (not irradiated)	89.5	89.7	90.6	89.9
ii.	Transmission (irradiated, 90 sec.)	37.1	22.8	21.1	21.1
iii.	Transmission (irradiated, 15 min.)	23.6	17.5	19.5	20.2
iv.	E, 90 sec	79.5	92.7	97.7	98.7
v.	Transmission (lightened, 2 min.)	35.6	45.1	56.6	70.2
vi.	Transmission (lightened, 10 min.)	58.1	75.5	85.3	86.3
vii.	A, 2 min.	18.2	38.2	52.2	71.7
viii.	A, 10 min.	52.4	80.3	92.5	94.8

20. Rows (i)-(iii) show the percent transmission for non-irradiated and irradiated samples. As seen in Row (i), prior to irradiation the percent transmission through the comparative samples is substantially equal to the percent transmission through the inventive samples. In the data shown in Rows

(ii)-(iii), the decrease in the percent transmission with the increase in irradiation time corresponds to a darkening of the samples.

21. Both the comparative samples and the inventive samples darken with exposure to irradiation, and the extent of darkening increases with longer exposure times.

22. The E value in Row (iii) of Table 1 provides a measure of the kinetics of the darkening. Specifically, the E value shows what percentage of the total measured darkening (after 15 minutes of irradiation) is achieved in the first 90 seconds of irradiation. Samples with a higher E value darken faster than samples with a lower E value.

23. After 90 seconds of irradiation, comparative samples 1 and 2 attain E values of 79.5 and 92.7%, respectively. In contrast, inventive samples 3 and 4 each attain E values greater than 97.5%. Thus, the additional formation of a second network (in Examples 3 and 4) results in a faster darkening compared to the single-resin samples.

24. Rows (v) and (vi) show the percent transmission for lightened samples after 2 and 10 minutes, respectively. Referring again to Table 1, after two minutes of lightening time, comparative samples 1 and 2 exhibit transmission values of just 18.2% and 38.2%, and after 10 minutes of lightening time, the transmission values of these samples are 58.1% and 75.5%, respectively. In contrast, inventive samples 3 and 4 exhibit transmission values of 56.6% and 70.2% after two minutes of lightening time, and 85.3% and 86.3%, respectively, after 10 minutes.

25. The A values in rows (vii) and (viii) of Table 1 shows what percentage of the total darkening (after 15 minutes of irradiation) is again lightened (after 2 minutes and 10 minutes). The A value is calculated as the ratio of the increase in transmission % (due to lightening) to the decrease in transmission % (due to darkening). Thus, samples with a higher A value recover (lighten) faster than samples with a lower A value.

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26. After 2 minutes of lightening time, the A values for comparative samples 1 and 2 are 18.2% and 38.2%, respectively, and after 10 minutes of lightening time, the A values for these samples are 52.2% and 71.7%. In contrast, after 2 minutes of lightening time, the A values for inventive samples 3 and 4 are 52.2% and 71.7%, and after 10 minutes of lightening time, the A values for the inventive samples are 92.5% and 94.8%. Surprisingly, the additional formation of a second network (in Examples 3 and 4) produces a faster lightening compared to the single-resin samples.

27. The superior photokinetic properties of the photochromic synthetic resin body produced according to the present invention are unexpected and surprising and could not have been expected or predicted based on the teachings of Sommerfeld et al., U.S. 5,868,101 and/or any other prior art of which I am aware.

28. Sommerfeld et al., U.S. Patent No. 5,886,101 does not teach that it would be possible to obtain an interpenetrating polymer network having a photochromic dye distributed homogeneously therethrough, or suggest the manufacture of a transparent synthetic resin body by producing first and second interpenetrating polymer networks and adding at least one photochromic dye before or during polymerization, and does not render the present invention obvious to a person skilled in the photochromic lens art.

29. All statements made herein of my own knowledge are true, and all statements made on information and belief are believed to be true, and further, these statements were made with the knowledge that willful statements and the like, so made, are punishable by fine or imprisonment, or both, under § 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the patent application or any patent issued thereon.

Date: 10.4.06

Respectfully Submitted,


Jobst Le Douss